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Uncertainty analysis on simple mass balance model to calculate critical loads for soil acidity

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A comprehensive uncertainty analysis, with advanced techniques and full list and full value ranges of all individual parameters, was used to examine a simple mass balance model and address questions of error partition and uncertainty reduction in critical acid load estimates that were not fully answered by previous studies.

Abstract

Simple mass balance equations (SMBE) of critical acid loads (CAL) in forest soil were developed to assess potential risks of air pollutants to ecosystems. However, to apply SMBE reliably at large scales, SMBE must be tested for adequacy and uncertainty. Our goal was to provide a detailed analysis of uncertainty in SMBE so that sound strategies for scaling up CAL estimates to the national scale could be developed. Specifically, we wanted to quantify CAL uncertainty under natural variability in 17 model parameters, and determine their relative contributions in predicting CAL. Results indicated that uncertainty in CAL came primarily from components of base cation weathering (BC_w; 49%) and acid neutralizing capacity (46%), whereas the most critical parameters were BC_w base rate (62%), soil depth (20%), and soil temperature (11%). Thus, improvements in estimates of these factors are crucial to reducing uncertainty and successfully scaling up SMBE for national assessments of CAL.
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Keywords: Critical load; Uncertainty analysis; Error partition; Sensitivity

1. Introduction

Development of effective policies for environmental protection requires timely and reliable information that can be used to assess the risks of pollutants on ecosystems at large spatial scales. The concept of a critical acid load (CAL hereafter) for forest soil is appealing because it provides simple, quantitative information about thresholds of pollutants over which unacceptable long-term harmful effects on ecosystem structure and function could occur (Nilsson and Grennfelt, 1988; Hodson and Langan, 1999; Gregor et al., 2004). The model of simple mass balance equations (SMBE) for CAL was developed and used in Europe for this purpose (de Vries, 1991; Sverdrup and De Vries, 1994; Werner and Spranger, 1996; Gregor et al., 2004). SMBE is simple in its model structure and data requirements and, thus, suitable for large-scale applications to identify potential areas of CAL exceedance. Other process-based models, like PROFILE (Sverdrup and De Vries, 1994; Hodson and Langan, 1999; Barkman and Alveteg, 2001; Hall et al., 2001) and MAGIC (Cosby et al., 2001; Gregor et al., 2004), may be more accurate at local scales and, therefore, more suitable for applications at particular sites where management decisions must be made, but pose great difficulty in scaling up beyond the watershed because they require intensive site-specific data that are often not available at large scales. However, to apply SMBE reliably and effectively for national assessments of CAL, the model must be tested for

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its adequacy by quantifying uncertainty in its predictions (Zak et al., 1997; Hodson and Langan, 1999; Barkman and Alveteg, 2001; Hall et al., 2001; Skeffington, 2006). Adequate uncertainty analysis is especially important for large scale modeling because it is not possible to validate spatial model predictions at large scales (Clark et al., 2001; Li and Wu, 2006).

Uncertainty analysis (UA) is the process for assessing uncertainty in modeling to: (1) identify major uncertainty sources; (2) quantify their degree and relative importance; (3) examine their effects on model predictions under different scenarios; and (4) determine prediction accuracy (O'Neill and Gardner, 1979; Petersen, 2000; Katz, 2002; Li and Wu, 2006). The importance of quantifying and reducing uncertainty in CAL estimates was recognized (Sverdrup and De Vries, 1994; Zak et al., 1997; Hodson and Langan, 1999; Barkman and Alveteg, 2001; Hall et al., 2001; Skeffington et al., 2006), and the previous work on uncertainty analysis of CAL estimates was examined in a comprehensive review by Skeffington (2006). However, even though many studies have provided insights into different aspects of uncertainty and ways of reducing uncertainty in CAL (Hodson and Langan, 1999; Barkman and Alveteg, 2001; Hall et al., 2001; Skeffington et al., 2006), systematic analysis of uncertainty in SMBE is still lacking. First, previous studies often focused on the mechanistic PROFILE model which is based on soil mineralogy (Hodson and Langan, 1999; Barkman and Alveteg, 2001). Therefore, uncertainty results from those studies could not be applied directly to SMBE. Second, previous studies primarily conducted sensitivity analysis (SA) in which model parameters were set at fixed percentages instead of using probability sampling techniques to define the parameter space (e.g., Jonsson et al., 1995; Zak et al., 1997; Barkman and Alveteg, 2001). Sensitivity analysis should not be used alone to address many of the uncertainty issues (e.g., defining the probability distribution for critical loads, quantifying error partition among factors) because SA and UA examine the same problem using different techniques that reveal different aspects of uncertainty (Li and Wu, 2006; see Section 2 for detailed discussion). Third, the simulations in previous studies were limited either by narrow data ranges because they were from a few sites or by incomplete analysis because they concentrated on parameter groups or selected parameters (Zak et al., 1997; Barkman and Alveteg, 2001). Without considering all individual factors, researchers could not fully assess the complexity of sensitivity and uncertainty in SMBE predictions of CAL. Fourth, the uncertainty measures used in some previous studies may be problematic. For example, use of the rank correlation coefficient lacked statistical power to quantify degree of importance by model parameters (e.g., Skeffington et al., 2006). Even though the general principles and techniques used by previous studies were well established, the methodology can and should be improved to provide better quantification and interpretation of uncertainty in CAL estimates as predicted by SBME.

The goal of this study was to improve our understanding of the behaviors of SMBE in terms of how natural variability in input data and model parameters impacts predictions of CAL. We decided not to perform uncertainty analysis of exceedance of CAL because exceedance is calculated as a simple linear function of CAL (Werner and Spranger, 1996; Gregor et al., 2004) and, as such, quantitative information about uncertainty of exceedance could be derived analytically from that of CAL (Li and Wu, 2006). We built upon previous studies (Hodson and Langan, 1999; Barkman and Alveteg, 2001; Hall et al., 2001; Skeffington et al., 2006), to conduct a more comprehensive assessment of uncertainty with three methodology improvements by: (1) examining all aspects of the SMBE behavior by treating the model as a mathematical construct; (2) considering all individual factors involved, the full ranges of their values, and their effects on both CAL and its key components; and (3) applying advanced techniques of uncertainty analysis so that the uncertainty of SMBE could be quantified and partitioned properly. This assessment should provide better and more detailed information that could be used to develop sound strategies for scaling up SMBE predictions of CAL with acceptable uncertainty to the national scale. Specifically, the objectives of this paper were to quantify the level of uncertainty in CAL predictions by SMBE, to determine the relative contributions of all individual factors to the total uncertainty in CAL, and to explore ways of reducing uncertainty for national assessments of CAL.

2. Methods

2.1. The SBME model and its factors

The SMBE model was developed in Europe for assessing potential risks of forest ecosystems to air pollutants (de Vries, 1991; Sverdrup and De Vries, 1994; Werner and Spranger, 1996; Gregor et al., 2004). The model assumed a simplified steady state with a single soil layer, was based on chemical criteria of biogeochemical processes, and had seven components that represented sources and sinks of soil acidity (Sverdrup and De Vries, 1994; Werner and Spranger, 1996; Watmough et al., 2004; Gregor et al., 2004). The SMBE model used in this study was expressed as

$$CAL(S+N) = BC_{dep} - CL_{dep} + BC_w - BC_u + N_i + N_u - ANC_{le,crit} \tag{1}$$

where CAL(S + N) is the critical loads for atmospheric deposition (wet and dry) of sulfur and nitrogen, BC_{dep} is base cation deposition, CL_{dep} is chloride deposition, BC_w is base cation weathering, BC_u is base cation uptake, N_i is nitrogen (N hereafter) immobilization, N_u is N uptake, and $ANC_{le,crit}$ is critical leaching of acid neutralizing capacity (Werner and Spranger, 1996; Watmough et al., 2004; Gregor et al., 2004). The unit of all the terms in Eq. (1) was eq ha^{-1} yr^{-1}, and the base cation (BC hereafter) was the sum of Ca, Mg, and K. Note that sulfur was not considered by SMBE because sulfur was assumed to be in equilibrium in soil solution (Gregor et al., 2004). Also, the original model had an eighth component, N denitrification (N_{de}), which was ignored in this study because N denitrification occurs within a relatively small proportion of the total land area (Watmough et al., 2004). Among the remaining seven components of CAL in Eq. (1), BC_w, BC_u, N_u, and $ANC_{le,crit}$ were referred to as key components of CAL and calculated by submodels with 14 important parameters, while BC_{dep} and CL_{dep} were defined by the input data from available GIS databases (McNulty et al., 2007) and N_i was defined by its range (Gregor et al., 2004; Table 1).

The submodels to calculate the four key components were defined as follows (de Vries, 1991; Sverdrup and De Vries, 1994; Werner and Spranger, 1996; Gregor et al., 2004). BC_w was based on the soil type-texture approximation method and expressed as

$$BC_w = R_{BC_w} \cdot (WR_C - 0.5) \cdot Z \cdot exp\left(\frac{A}{281} - \frac{A}{273+T}\right) \tag{2}$$

where R_{BC_w} is BC weathering rate per unit depth of soil, WR_c is weathering rate class, Z is soil depth, T is average annual air temperature, and A is the

Table 1
The data (means, standard deviations, ranges) of the factors used in uncertainty analysis of SMBE

Factor	Symbol	Mean	SD [Range]	Unit	Distribution
BC weathering	BC_w				
Soil depth[a]	Z	1.23	0.38	m	Normal
Temperature[a]	T	11.90	5.19	°C	Normal
BC_w base rate[b]	R_{BC_w}	750.0	[225,2250]	eq ha^{-1} yr^{-1} m^{-1}	Triangular
Uptake of BC and N[c]	BC_u, N_u				
Growth rate	K_{gr}	9.03	3.68	m^3 ha^{-1} yr^{-1}	Normal
Stem wood density	ρ_{st}	551.67	89.42	kg m^{-3}	Normal
Branch to stem ratio	$f_{br,st}$	0.18	0.07	kg kg^{-1}	Normal
BC content in stem	$ctBC_{st}$	0.13	0.03	eq kg^{-1}	Normal
BC content in branch	$ctBC_{br}$	0.26	0.05	eq kg^{-1}	Normal
N content in stem	ctN_{st}	0.11	0.03	eq kg^{-1}	Normal
N content in branch	ctN_{br}	0.35	0.08	eq kg^{-1}	Normal
Acid neutralizing capacity	$ANC_{le,crit}$				
Runoff[a]	Q	391.27	507.04	m^3 ha^{-1} yr^{-1}	Normal
Gibbsite constant[d]	K_{Gibb}	500.00	300.00	m^6 eq^{-2}	Log-Normal
BC:Al ratio[e]	$R_{BC:Al}$	5.5	[1,10]	mol mol^{-1}	Triangular
BC_w percent[f]	P_{le}	77.50	[65,90]	%	Uniform
Input CL component					
N immobilization[g]	N_i	42.85	[14.3,71.4]	eq ha^{-1} yr^{-1}	Triangular
BC deposition[h]	BC_{dep}	136.97	87.07	eq ha^{-1} yr^{-1}	Log-Normal
Chloride deposition[h]	CL_{dep}	52.96	68.08	eq ha^{-1} yr^{-1}	Log-Normal

There were 14 parameters for calculation of the four key components and three input components of critical loads. The values and distributions of the parameters were primarily based on literature (de Vries et al., 1993; Sverdrup and De Vries, 1994; Werner and Spranger, 1996; Barkman and Alveteg, 2001; Hall et al., 2001; Gregor et al., 2004; Skeffington et al., 2006) with some statistics from GIS databases (McNulty et al., 2007). SD stands for standard deviation, BC for base cation, and N for nitrogen.

[a] From spatial databases of environmental variables (Daly et al., 1994; Miller and White, 1998; McNulty et al., 2007).

[b] From the relation of $R_{BC_w}(WR_C - 0.5)$ by assuming that BC_w class range from 1–5 with the average class of 2 (Werner and Spranger, 1996; Gregor et al., 2004).

[c] From information about species-specific data of five coniferous and seven deciduous trees (de Vries, 1991; Werner and Spranger, 1996; Gregor et al., 2004).

[d] From the ranges given in Gregor et al. (2004); (Werner and Spranger, 1996).

[e] from the commonly-used values of 1.0 for Europe (Sverdrup and De Vries, 1994; Hall et al., 2001) and 10.0 for Canada (Watmough et al., 2004; Ouimet et al., 2006).

[f] From the ranges given in Gregor et al. (2004; Page V23) as functions of soil types (ranging from 70% for poor sandy soils to 85% for rich soils).

[g] From values given in Gregor et al. (2004, page V13).

[h] From spatial databases of wet deposition (Grimm and Lynch, 2004; McNulty et al., 2007).

Arrhenius constant (de Vries et al., 1993; Werner and Spranger, 1996; Gregor et al., 2004). Z and T were defined by GIS databases (McNulty et al., 2007; Table 1). A was based on the Arrhenius relation and set at 3600 K (Sverdrup and De Vries, 1994; Werner and Spranger, 1996; Gregor et al., 2004).

The key component of BC_w defined in Eq. (2) was a function of both R_{BC_w} and WR_c, i.e., $R_{BC_w}(wR_C - 0.5)$, with correction factors for soil depth and for using air temperature as a measure of soil temperature. R_{BC_w} was treated in Gregor et al. (2004) as a constant (500 eq ha^{-1} yr^{-1} m^{-1}), even though R_{BC_w} was the parameter related directly to BC_w. Thus, BC_w defined by Eq. (2) was controlled primarily by WR_c as a function of soil texture and parent material with classes of 1–6 (de Vries et al., 1993; Sverdrup and De Vries, 1994; Werner and Spranger, 1996; Gregor et al., 2004; Table 1). The BC_w model in Eq. (2) allowed for easy parameterization and scaling of site level data to regional or national scales. However, preliminary analyses suggested that WR_c was one of the most critical factors in SMBE predictions of CAL. As a result, the uncertainty analysis of SMBE should be enhanced when the base rate of BC_w was represented by a numerical variable (R_{BC_w}) instead of a categorical variable (WR_c). Therefore, Eq. (2) was reformulated as

$$BC_w = R_{BC_w} \cdot Z \cdot exp\left(\frac{A}{281} - \frac{A}{273+T}\right) \tag{3}$$

In the modified model (Hodson and Langan, 1999), R_{BC_w} represented the BC_w base rate per depth of soil, but changed as a variable based on conditions of soil texture and parent material (Gregor et al., 2004). R_{BC_w} was assumed to range from 225 to 2250 (eq ha^{-1} yr^{-1} m^{-1}) based on the given range of WR_c from 1 to 5 (assuming class 6 to be rare; Gregor et al., 2004; Table 1). Note that the results of R_{BC_w} could be translated back to WR_c if needed for direct comparison.

The two uptake components (BC_u, N_u) shared the same formula, which was expressed as

$$Y_u = K_{gr} \times \rho_{st} \cdot \left(ctY_{st} + f_{br,st} \cdot ctY_{br}\right) \tag{4}$$

where Y_u is either BC_u for BC uptake or N_u for N uptake, K_{gr} is average annual growth rate, ρ_{st} is density of stem wood, $f_{br,st}$ is branch-to-stems ratio, ctY_{st} is content in stems for BC or N, and ctY_{br} is content in branches for BC or N (Gregor et al., 2004). These seven uptake-related parameters in Eq. (4) were functions of tree species (de Vries, 1991; Werner and Spranger, 1996; Gregor et al., 2004; Table 1). BC_u and N_u represented long-term average removal of these elements from ecosystems and would be subject to harvesting methods used (Werner and Spranger, 1996; Gregor et al., 2004).

$ANC_{le,crit}$ was based on the critical aluminum concentration method and expressed as

$$ANC_{le,crit} = -Q^{2/3} \cdot \left[1.5 \frac{BC_{dep} + P_{le} \cdot BC_w - BC_u}{K_{Gibb} \cdot R_{BC:Al}}\right]^{1/3} - 1.5 \frac{BC_{dep} + P_{le} \cdot BC_w - BC_u}{R_{BC:Al}} \tag{5}$$

where Q is annual runoff, P_{le} is percent of BC_w involved in leaching, $R_{BC:Al}$ is ratio of BC to aluminum, and K_{Gibb} is the Gibbsite constant (Werner and

Spranger, 1996; Gregor et al., 2004). Q and BC_{dep} were defined by GIS databases (McNulty et al., 2007; Table 1), while BC_w and BC_u were calculated by Eqs. (3) and (4). $R_{BC:Al}$ and P_{le} were functions of soil type, and K_{Gibb} was a function of soil type and organic material (Werner and Spranger, 1996; Gregor et al., 2004). It should be pointed out that P_{le} was not in the original model of $ANC_{le,crit}$, but was discussed in Gregor et al. (2004) and Sverdrup and de Vries (1994) as a factor to determine different BC_w values that may be considered in calculating $ANC_{le,crit}$ based on soil types. It was included here in Eq. (5) because of its importance to $ANC_{le,crit}$ as suggested by preliminary simulation results. In addition, on the right side of Eq. (5), the first term represented hydrogen ion concentration, $[H^+]$, and the second term represented inorganic aluminum concentration, $[Al^{3+}]$ (Sverdrup and De Vries, 1994; Gregor et al., 2004; Skeffington, 2006).

2.2. The data used in simulations

The statistical values of all factors (i.e., 3 components and 14 parameters) used in the uncertainty analysis of SMBE are given in Table 1. Some values were averages and standard deviations (SD) obtained from spatial databases (e.g., those for soil depth, temperature, runoff, BC deposition, and chloride deposition) (Miller and White, 1998; Grimm and Lynch, 2004; McNulty et al., 2007). Many other factors were known only for their ranges of variability, such as BC_w base rate, BC:Al ratio, BC_w percent, and N immobilization (Werner and Spranger, 1996; Gregor et al., 2004). In addition, some parameters were based on the relationships developed for SMBE (e.g., the Gibbsite constant and the uptake-related parameters; de Vries et al., 1993; Sverdrup and De Vries, 1994; Werner and Spranger, 1996; Gregor et al., 2004). In this study, we assumed that: (1) the environmental variables and the uptake variables with known mean and SD should have normal distributions; (2) the deposition variables and the Gibbsite constant have lognormal distributions; (3) the interval variables only with known ranges have triangular distributions (whose expected values were the mid values of the ranges when unknown); and (4) the categorical variables (e.g., BC_w percent) have uniform distributions (Table 1). Similar assumptions about data distributions have been cited in other studies (Hodson and Langan, 1999; Barkman and Alveteg, 2001; Hall et al., 2001; Skeffington et al., 2006).

2.3. Approaches to uncertainty quantification

The most common approach to quantitative assessment of uncertainty in model predictions is to run the model under perturbations (i.e., changing values of parameters and input data) with the help of Monte Carlo simulations (MCS) (Gardner and O'Neil, 1983; Gardner et al., 1990; Rastetter et al., 1992; Heuvelink, 1998; Jansen, 1998; Katz, 2002; Li and Wu, 2006; Skeffington, 2006). The specific questions about uncertainty in CAL estimation with SBME were: Which was the most critical factor to the prediction and uncertainty of CAL? What was the relative contribution by each factor to the model uncertainty? What were the general patterns of SBME behaviors that may be used to develop strategies of improving model accuracy and reducing prediction uncertainty in the scaling up process? To address these questions, different simulation strategies were needed.

2.3.1. Sensitivity analysis

To determine the most critical factors to SMBE predictions of forest soil CAL, we used the technique of single-parameter sensitivity analysis (SA) to rank parameters based on the rates of change in model output caused by changes in the values of a particular parameter (Klepper, 1997; Katz, 2002; Li and Wu, 2006; Skeffington, 2006). The SA procedure was: (1) to set all parameters to their average (or most likely) values, (2) to change one parameter at a time by a given percentage (e.g., a reduction or increase by 10%, 20%, 30%), (3) to run SMBE to estimate CAL for each parameter set (which was labeled as simulation MCS_{SA}), and (4) to assess the sensitivity of CAL estimates to the parameters by a measure of relative error of each simulation (Saltelli et al., 2000; Melching and Bauwens, 2001; Katz, 2002; Li and Wu, 2006). The measure of relative error was defined for a given factor by

$$RE_{y\%} = \left(V_{y\%} - V_{100\%}\right)/V_{100\%} \tag{6}$$

where $RE_{y\%}$ is the index of relative error in predicted variable V (i.e., percent change in model output caused by a fixed percent change in a model input), $V_{100\%}$ is the value of the predicted variable obtained from the simulation in which all factors were set at average values (i.e., 100%; Table 1), and $V_{y\%}$ is the value of the predicted variable obtained from the simulation in which the target factor was changed to a certain (y) percentage of its average value while all the other factors were kept at the average values. The predicted variable V could be either CAL or one of the key components. Thus, RE was used to quantify the sensitivity ranking of CAL to a factor. The higher the RE value, the more sensitive the CAL estimate was to the factor. SA was used to describe the effects of model parameters on the estimates of CAL.

2.3.2. Uncertainty analysis

To determine the relative contributions of all parameters to uncertainty of the SMBE predictions of forest soil CAL, we used the uncertainty analysis (UA) technique to partition variability in CAL estimates among the considered parameters (Jansen, 1998; Katz, 2002; Li and Wu, 2006). The UA procedure was: (1) to use the Latin hypercube sampling (see below) to select a number of values for each parameter based on its assumed probability distribution, (2) to define the parameter space by a full factorial of all sample values of the parameters, (3) to run SMBE with all of the parameter sets in the parameter space to calculate the total variability of the model output (which was labeled as simulation MCS_{ALL}), (4) to run SMBE again, with only the parameter sets in which a target parameter X was fixed at its average value while all other parameters were allowed to change at full ranges of their values, to calculate the marginal variance of the target parameter X (which was labeled as simulation MCS_{ALL-X}), and (5) to quantify the error contribution of the parameter based on the proportion of its marginal variance in the total variability of the model output (Eq. 7; Katz, 2002; Li and Wu, 2006). The error contribution of any given parameter (X) was calculated by

$$E_X = \frac{\left(\sigma^2_{ALL} - \sigma^2_{ALL-X}\right)}{\sigma^2_{ALL}} \tag{7}$$

where E_X is the index of error contribution by factor X, σ^2_{ALL} is the overall variance of the model output obtained from the all-factor simulation MCS_{ALL}, and σ^2_{ALL-X} is the top-marginal variance of the model output obtained from the all-but-one-factor simulation MCS_{ALL-X}. Because σ^2_{ALL-X} reflects the variability reduced by the absence of the target factor, E_X can be used to define the upper limits of potential error reduction and determine the error partition of CAL among the selected parameters. UA was used to quantify the effects of model parameters on the variability of CAL.

2.3.3. Sampling

The Latin hypercube sampling was used to obtain a representative sample of parameter values and to reduce the computational burden (McKay et al., 1979; Li and Wu, 2006). The sampling was done with the statistics and the assumed distributions of all the parameters involved (Table 1). In the Latin hypercube sampling, the range of a parameter was stratified into k equal probability segments based on the assumed theoretical probability distribution (Table 1), and each stratum was randomly sampled once. The choice of k sample values for each parameter was made to balance the computational limitation and the need to have a sample size large enough to represent a full range of the parameter values, given the complexity of the model involved. In the current study, three sample values for each factor were used after preliminary results showed that they produced patterns similar to simulations with ten sample values. This small sample was primarily dictated by the computational limitation because the parameter space as defined by the full factorial of the three values from all 17 factors already formed a huge domain (see below).

2.3.4. Simulation strategies

Four analyses were performed to provide a full assessment of uncertainty in modeling CAL with SMBE. Each analysis ran the simulations of MCS_{SA}, MCS_{ALL}, and MCS_{ALL-X} with a different group of parameters.

1. The *first* analysis was used to quantify uncertainty in CAL by the seven components as targeted factors (Eq. 1). All of the 17 parameters (Table 1) were used in the simulations, but only the variability of the components

was considered in calculating the uncertainty measures of $RE_{y\%}$ and E_X (Eqs. 6 and 7).

2. The *second* analysis was used to assess relative importance of all individual parameters to CAL in SMBE. The targeted factors included the 14 parameters and the three input components involved in Equations 1, 3, 4, and 5 (Table 1).
3. The *third* analysis was used to examine uncertainty in the intermediate outputs of SMBE (i.e., the four modeled components of BC_w, BC_u, N_u, and $ANC_{le,crit}$). For each key component, all parameters involved (Eqs. 3, 4, or 5) were used to assess sensitivity and uncertainty in CAL and the component of interest predicted by SMBE.
4. The *fourth* analysis was used to highlight effects of using extreme values of BC:Al ratio (Eq. 5) on outputs of uncertainty analysis, keeping everything else the same as those in the *second* analysis. Instead of the probabilistic samples, the minimum (1.0, a common value used for Europe; Sverdrup and De Vries, 1994; Hall et al., 2001), the maximum (10.0, a common value used for Canada; Watmough et al., 2004; Ouimet et al., 2006), and the average of BC:Al ratio were used in simulations (Table 1). BC:Al ratio was selected for this particular analysis because it was the only factor that showed significant impact with the extreme values in preliminary analysis, and is the only environmental factor in SMBE that defines potential damage to ecosystems (Hodson and Langan, 1999).

For MCS_{SA} of each analysis, SMBE was run with seven parameter sets for each factor (i.e., a reduction or increase by 0%, 10%, 20%, and 30% of its average value). For MCS_{ALL} and MCS_{ALL-X} of each analysis, the numbers of simulations were 3^N and 3^{N-1} (or 3^{N-k} if the targeted factor was a modeled key component with itself having k parameters), depending on the number (N) of parameters examined. For example, the total number of parameter sets used in MCS_{ALL} to examine effects of all 17 factors on CAL (i.e., in the first and the second analyses) was 129,140,163 (i.e., 3^{17}). The large number of simulations was achieved by embedding the UA routines (including sampling algorithms) inside SMBE. The summation of the E_X values (Eq. 7) by all parameters may exceed 100% because of the complexity (e.g., nonlinearity) in the submodels of SMBE. Thus, the calculated E_X values were standardized to highlight the relative importance of each factor involved (i.e., the reported E_X values were forced to sum to 100%).

3. Results

3.1. Sensitivity and uncertainty of CAL: key components

The results from the first simulation analysis under a full range of the parameter space are displayed in Tables 2 and 3, and Figs. 1 and 2. The average of forest soil CAL was estimated at 1887 eq ha^{-1} yr^{-1} with a median of 1765 and a SD of 770. The 5% and 95% quantiles were 864 and 3607 eq ha^{-1} yr^{-1} (Table 2). In addition, the results were summarized to present the frequency distribution of CAL (Fig. 1) and the cumulative distribution functions of CAL and the three key components (Fig. 2). The seven components used in SMBE differentially impacted the predicted CAL (Table 3). Among the components, CAL was the most sensitive to BC_w followed by N_u and BC_u. For example, a 20% change in BC_w caused a 17% change in CAL estimates (i.e., $RE_{20\%}$ in Table 3). In contrast, BC_w and $ANC_{le,crit}$ were the most influential to uncertainty of CAL, contributing 49% and 46% of variability to the total error in CAL estimates, respectively (Table 3).

3.2. Sensitivity and uncertainty of CAL: all parameters

The results from the second simulation analysis are displayed in Fig. 3 to examine effects on CAL by all 17 of the individual factors in SMBE (Table 1). CAL sensitivity was influenced the most by BC_w base rate, soil depth, temperature, BC content in stems, and N content in stems (Fig. 3A). With a 20% increase of these factors, CAL estimates showed a 22% increase by BC_w base rate and soil depth, 12% by temperature, and 8% by N content in stems, but a 13% decrease by BC content in stems. Variation in all other factors had little impact on CAL, except for growth rate, wood density, and BC_w percent, each of which varied CAL by just under 5%. CAL uncertainty came primarily from three factors: BC_w base rate (62%), soil depth (20%), and temperature (11%) (Fig. 3B). No other factors contributed more than 3% of the error to forest soil CAL.

3.3. Sensitivity and uncertainty of key components: individual parameters

The results of effects of individual parameters on the corresponding key components (Eqs. 3–5) are summarized in Table 4, and Figs. 4 and 5. For sensitivity, the most influential parameters were BC_w base rate and soil depth to BC_w, and growth rate and stem wood density to BC_u (Table 4, Fig. 4A and B). Note that N_u had almost identical sensitivity values to those of BC_u. The most critical parameters controlling $ANC_{le,crit}$ were growth rate, stem wood density, BC_w base rate, soil depth, BC_w percent, and BC content in stems (Table 4, Fig. 4C). The critical parameters to BC_w and BC_u displayed straight proportional effects on these corresponding components (Fig. 4A and 4B). However, the parameters critical to

Table 2
The statistical values (i.e., mean, SD, range, median) of the seven components from the first simulation analysis

Component	Mean	SD	Minimum	Maximum	Median
BC weathering	1571.92	755.95	665.58	3140.05	1492.63
BC uptake	837.21	335.54	360.74	1615.29	810.50
N uptake	828.74	270.56	347.78	1622.00	813.30
Acid neutralizing capacity	−192.52	168.54	−1045.57	−0.28	−163.36
N immobilization	43.52	10.71			
BC deposition	130.83	65.07			
Chloride deposition	43.06	34.90			
Output: critical loads	1887.25	770.39	187.74	4656.87	1765.33

The values for critical loads, BC_w, BC_u, N_u and $ANC_{le,crit}$ were obtained from simulations MCS_{ALL}, while those for N_i, BC_{dep}, and CL_{dep} were from input data (Table 1). The 5% and 95% quantiles of critical loads were also measured at 864.06 and 3606.7. All variables were in the unit of eq ha^{-1} yr^{-1}.

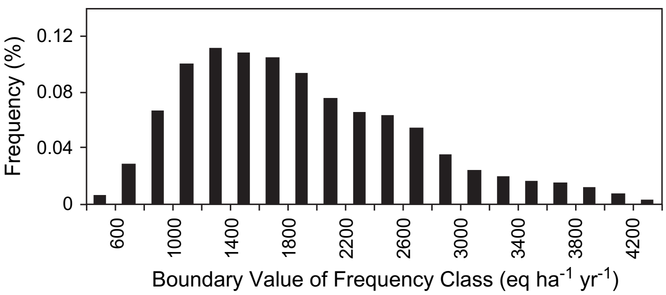

Fig. 1. Frequency distribution of critical loads from the first simulation analysis under a full range of the parameter space based on the probability sampling. The figure shows a positively skewed distribution (average at 1887 and median at 1765 eq ha^{-1} yr^{-1}), which is similar to a log-normal distribution.

$ANC_{le,crit}$ showed compounded effects of the parameters with a 20% increase of the critical parameters leading to changes of 82–98% in $ANC_{le,crit}$ values (Table 4, Fig. 4C). In addition, the BC:Al ratio and Gibbsite constant exerted different effects on $ANC_{le,crit}$ (Fig. 5). For uncertainty, the results identified critical parameters with the most error contributions to the key components (Table 4). For predictions of BC_w, all three parameters were important, with BC_w base rate contributing 74% and soil depth 19%. For BC_u, the parameters that contributed the most were growth rate (75%), followed by stem wood density (11%) and BC content in stems (11%). Patterns similar to BC_u were observed for N_u

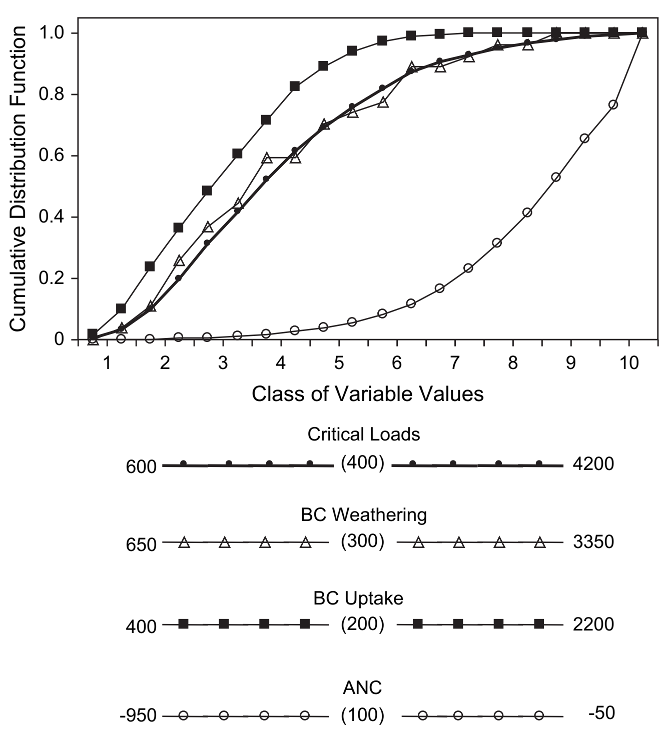

Fig. 2. Cumulative distribution functions of critical loads and three key components. The data were from MCS_{ALL} of the first simulation analysis. Note that the distribution of N_u was omitted from the figure because it was almost identical to that of BC_u. The statistics of these predicted variables were given in Table 2. The values of these variables corresponded to the classes were marked by the legends with increment values shown in the parentheses. For example, for critical loads (eq ha^{-1} yr^{-1}), class 1 corresponded to 600 and class 10 to 4200 with the increment being 400.

Table 3
Relative importance of the seven components to uncertainty in critical loads based on the first simulation analysis

Component	$RE_{20\%}$ (%)	E_X (%)
BC weathering	16.66	49.40
BC uptake	−8.87	2.00
N uptake	8.78	2.00
Acid neutralizing capacity	2.04	46.17
N immobilization	0.67	0.01
BC deposition	2.85	0.34
Chloride deposition	−0.83	0.07

The values of the relative error ($RE_{20\%}$; Eq. 6) were obtained from simulations MCS_{SA}, representing relative change in critical load estimates when a given target component was increased by 20%. The values of the error contribution (E_X; Eq. 7) were obtained from simulations MCS_{ALL} and MCS_{ALL-X}, defining how the error in the critical loads estimates was partitioned among the components.

with slight differences in the percentages. For predictions of $ANC_{le,crit}$, the most critical parameters were BC_w base rate (57%), soil depth (18%), temperature (10%), and growth rate (9%), respectively.

3.4. Changes in sensitivity and uncertainty of CAL: extreme values of BC:Al ratio

The results from the fourth simulation analysis are highlighted for eight selected factors in Table 5. BC:Al ratio was the factor of interest, while the other seven parameters were selected because they had highest uncertainty rankings as observed in Fig. 3. The main contributors to uncertainty of CAL and $ANC_{le,crit}$ were in similar orders and with the top three ranked parameters being: BC_w base rate (48% and 45%), BC:Al ratio (29% and 28%), and soil depth (14% and 13%).

4. Discussion

4.1. Effects of key components on critical loads

Uncertainty of CAL as predicted by SMBE under a full range of the probabilistic parameter space was at a medium level, with SD of 770 eq ha^{-1} yr^{-1} and CV of 40% (Table 2). The standard deviation of CAL varies when parameter variability changes and when parameters are added to or removed from the SD calculation. For example, SD of CAL increased greatly to 3345 eq ha^{-1} yr^{-1} (CV at 150%) when extreme parameter values (i.e., minimum, average, and maximum) were used in a preliminary simulation analysis. In particular, the use of the extreme values of BC:Al ratio alone increased the CV of CAL from 40% to 50% ($MCS_{ALL}$of the second analysis vs. MCS_{ALL} of the fourth analysis). For removal of single parameters (e.g., MCS_{ALL-X} of the second simulation analysis), CV of CAL could get as low as 28% when BC_w base rate was kept at its average value. The statistical values of CAL obtained in this study were similar to those from the previous studies cited in Skeffington (2006), but the means and medians showed relatively higher values (e.g., Table 2). Nevertheless, accurate predictions of CAL were not the concern of this study; our primary focus was on the uncertainty of CAL and

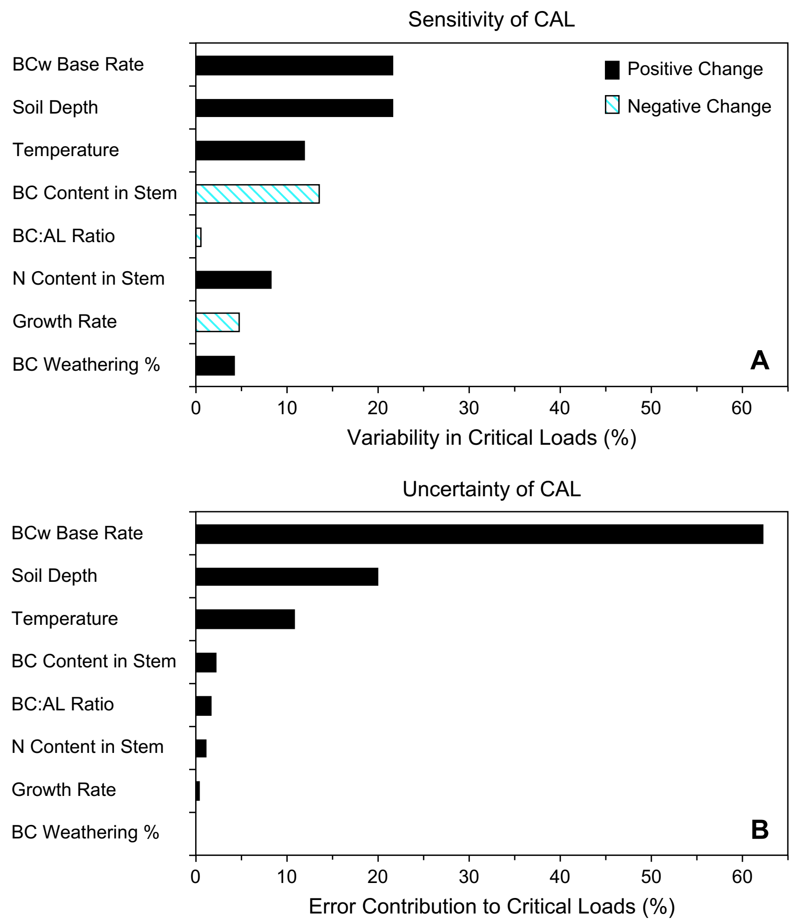

Fig. 3. Most critical factors to the sensitivity and uncertainty of critical load estimates. The data were from the second simulation analysis. Similar to Table 3, the figure shows the ranking of (A) sensitivity of critical load (i.e., $RE_{20\%}$ defined in Eq. 6) and (B) uncertainty of critical load (i.e., E_X calculated by Eq. 7). Note that the factors were chosen primarily for their uncertainty ranking, except for the last two factors (i.e., growth rate, BC_w percent). Factors not shown in the figure had insignificant effects on uncertainty of critical loads, but a few (i.e., wood density, BC in branch, N in branch) had $RE_{20\%}$ values similar to those of growth rate and BC_w percent at below 5%.

its most critical contributors that were revealed by treating SMBE as a mathematical construct in a systematic uncertainty analysis. Given the degrees of variability among the factors used in SMBE (Table 1), the SD of CAL observed with the full probabilistic parameter space suggests that uncertainty from SMBE and its parameters may be moderate in the scaling up of CAL.

The results indicated that much of the uncertainty in CAL as predicted by SMBE came from BC_w and $ANC_{le,crit}$ with each respectively contributing 49% and 46% to the total variability in CAL estimates (Table 3). Even though they have been identified as the most critical components before (Hodson and Langan, 1999; Hall et al., 2001; Skeffington, 2006), the results from our study quantitatively demonstrate the dominance of BC_w and $ANC_{le,crit}$ in SMBE estimates of CAL. Our results further indicate that, despite their similar contributions to the uncertainty of CAL, BC_w may be more important because the most critical factors to $ANC_{le,crit}$ were also the three parameters of BC_w (Table 4). However, one must be cautious and not overestimate the potential of error reduction that may be achieved through improved estimates of BC_w. How much of the 49% of CAL variability accounted for by BC_w may be removed is unknown because natural variability in BC_w and its parameters as major sources of uncertainty cannot be eliminated (Li and Wu, 2006). Nevertheless, the best way to reduce uncertainty in CAL estimates should be to improve the accuracy of BC_w estimates.

The five other components showed insignificant contributions to variability of CAL estimates (Table 3). For BC_u and N_u, the reason may be that they offset each other in the estimation of CAL because BC_u acted as a source of soil acidity while N_u functioned as a sink (Sverdrup and De Vries, 1994; Gregor et al., 2004). This relationship is reasonable given that these uptake components tend to vary together as timber harvesting removes both BC_u and N_u from the system. However, if BC_u and N_u were removed in disproportion to input levels, then harvesting could significantly impact nutrient balance. For N_i, BC_{dep}, and CL_{dep}, the reason may be that these input components to CAL were observations and therefore had less uncertainty. Nevertheless, these components of CAL should not be discounted entirely because they directly affect CAL estimation.

The results from simulations under the full range of the probabilistic parameter space also suggested that CAL follow

Table 4
Relative importance of model parameters to the uncertainty in four key components of critical loads

Component	Factor	$RE_{20\%}$ (%)	E_X (%)
BC weathering	BC_w base rate	20.00	74.41
	Soil depth	20.00	18.79
	Temperature	11.03	6.80
BC uptake	Growth rate	20.00	74.77
	BC content in stem	14.53	11.31
	Stem wood density	20.00	11.23
	Branch to stem ratio	5.47	2.40
	BC content in branch	5.47	0.29
N uptake	Growth rate	20.00	71.25
	Stem wood density	20.00	12.33
	N content in stem	12.53	9.73
	Branch to stem ratio	7.47	5.72
	N content in branch	7.47	0.98
Acid neutralizing capacity	BC_w base rate	91.84	56.78
	Soil depth	91.84	17.89
	Temperature	51.48	9.73
	Growth rate	−98.29	9.07
	BC:Al ratio	−12.45	1.65
	BC content in stem	−82.21	1.64
	Stem wood density	−98.29	1.59
	BC deposition	15.26	0.45
	Branch to stem ratio	−27.06	0.44
	Runoff	4.61	0.30
	Gibbsite constant	−2.28	0.22
	BC content in branch	−27.06	0.16
	BC_w percent	91.84	0.089

The data were from the third simulation analysis. The uncertainty measures, $RE_{20\%}$ and E_X, were the same as those described in Table 3. The factors were arranged based on their uncertainty rankings.

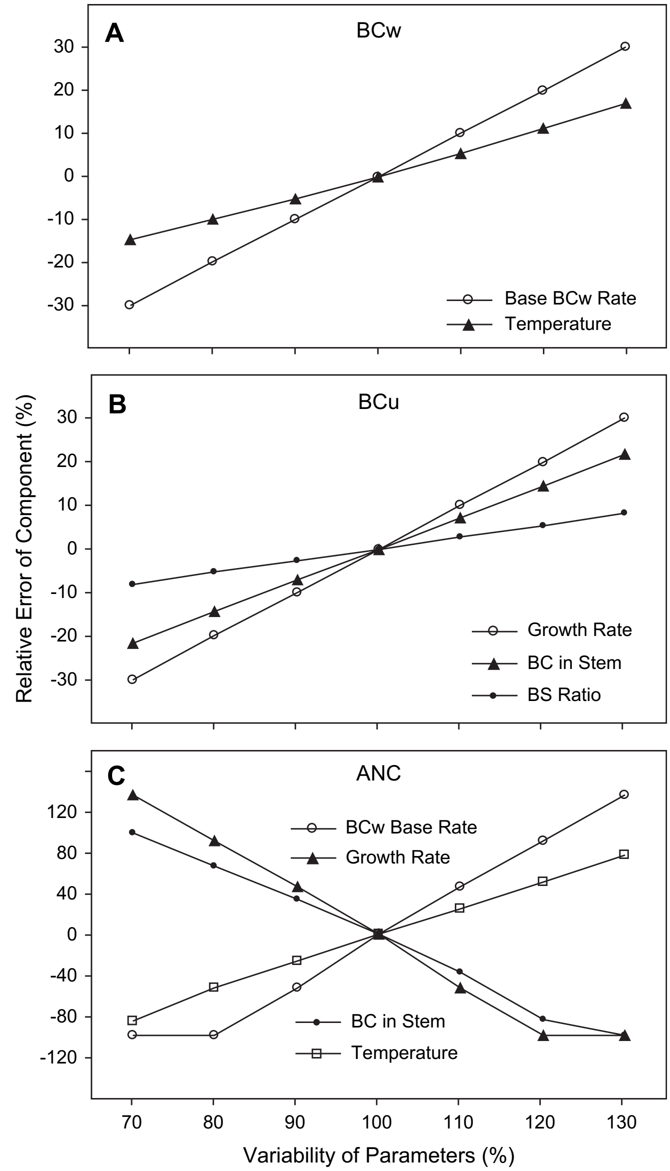

Fig. 4. Sensitivity of the three key components to individual parameters based on the third simulation analysis. Note that the sensitivity curves of N_u were omitted from the figure because they were almost identical to those of BC_u. Each panel was set up similarly; the index of relative error (RE; Eq. 6) represented the percent change in each key component caused by a fixed percent change in a corresponding input parameter. Note that not all parameters were shown in the figure. For BC_w, soil depth was not displayed because it had the identical sensitivity curve with BC_w base rate. For BC_u, density of stem wood and BC in branches were not displayed because they had the identical sensitivity curves with growth rate and branch to stem ratio, respectively. For $ANC_{le,crit}$, three parameters were also important but not displayed because soil depth and BC_w percent had the identical sensitivity curves with BC_w base rate, while density of stem wood had the same sensitivity with growth rate. All of the other seven parameters were insignificant (e.g., having relative errors less than 28% when they were changed by 20%; see Table 4).

a log-normal distribution (Fig. 1). The log-normal distribution may be characteristic for CAL because SMBE outputs from both simulations reported here and preliminary analyses indicated that the average and SD of CAL may change as different subsets of parameters were used, but the distribution remained the same. The distribution of CAL may have resulted primarily from the log-normal distribution of BC_w, because of the various statistical distributions observed (Fig. 2) for the four key components and assumed for the individual parameters (Table 1). The frequency distribution of CAL obtained here from the probabilistic samples of parameters should have a stronger statistical basis than those obtained from the non-probabilistic samples. Therefore, the confidence interval (CI) calculated based on a normal distribution should not be used to define the uncertainty level of CAL (e.g., Barkman et al., 1995; Barkman and Alveteg, 2001) because for asymmetric probability distributions such as log-normal, CI is also asymmetric (Meeker and Escobar, 1998). For example, the 5% and 95% quantiles for CAL were 864 and 3606 eq ha^{-1} yr^{-1} (Table 2). However, if normality were assumed for CAL, then the calculated 5% and 95% quantiles would be 619 and 3154 eq ha^{-1} yr^{-1}. Thus, it is imperative that, before parametric analyses like CI can be used, the distribution of a model output should be determined to avoid false projections.

4.2. Effects of individual parameters on critical loads

The variability and uncertainty in forest soil CAL estimates were controlled primarily by five parameters in SMBE (i.e., BC_w base rate, temperature, soil depth, BC content in stems, and N content in stems; Fig. 3). Among them, BC_w base

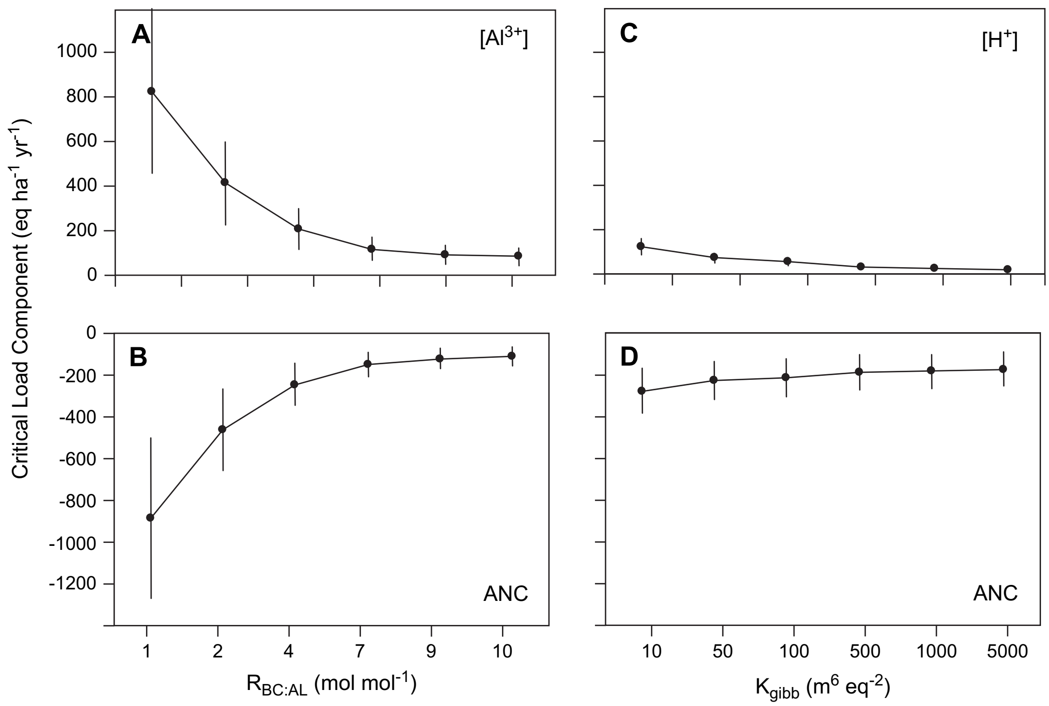

Fig. 5. Relationships of $ANC_{le,crit}$, $[H^+]$, and $[Al^{3+}]$ to $R_{BC/Al}$ and K_{Gibb}, two parameters related to the chemical criterion of critical loads (Gregor et al., 2004; Skeffington, 2006). Note that $[H^+]$ and $[Al^{3+}]$ were the two components of $ANC_{le,crit}$ in Eq. (5). Values in the figure were averages (circles) with one SD (bars).

rate, soil depth, and temperature (i.e., the three parameters of BC_w) caused the most change and contributed the highest uncertainty (i.e., 93% combined) in CAL estimates (Fig. 3). The significance of these results lies in the in-depth and systematic analyses of our approach because all 17 factors in SMBE were considered individually and together for their effects on CAL. Barkman and Alveteg (2001) speculated that analysis with all input parameters should greatly improve quantification of uncertainty. Such information on error partition among the parameters, as well as among the components, provides the direct indication of how uncertainty in CAL may be reduced in any large scale assessment of CAL with SMBE.

4.3. Effects of parameters on key components

The four modeled components of SMBE (i.e., BC_w, $ANC_{le,crit}$, BC_u, N_u) were treated as the intermediate model outputs and examined separately for their own uncertainty in the submodels (Table 4). The results from a closer analysis on the key components can provide additional insights into the behaviors of SMBE.

For BC_w, all three parameters were important. Sensitivity of BC_w was high to BC_w base rate and soil depth, each of which led to proportional change in BC_w (Figs. 3 and 4A, Table 4). Uncertainty in BC_w was dominated by BC_w base rate (74%), but soil depth and temperature also made considerable contributions (Fig. 3, Table 4). Barkman and Alveteg (2001) found that soil physical properties were the most important predictors of BC_w. Our results support their finding because BC_w base rate is closely related to soil type and physical properties. However, unlike the PROFILE model examined by Barkman and Alveteg (2001), no difference in SMBE predictions were found between CAL and BC_w in terms of the ranking of importance of identified parameters.

For BC_u and N_u, the most influential parameters were growth rate, stem wood density, and BC or N contents in stems (Fig. 4B, Table 4). Growth rate and stem wood density caused proportional changes in the two uptake components because these parameters were used in multiplicity in the uptake submodel (Eq. 4). Growth rate was the primary parameter of uncertainty in BC_u and N_u with E_X values over 71%, but stem wood density and BC or N content in stems also showed some contributions in predicting BC_u and N_u (Table 4). These results indicate that there were significant differences between CAL and the uptake components in terms of the ranking of the parameters because BC and N contents in stems were more important parameters to CAL than growth rate and stem wood density (Fig. 3). It is not clear why this disparity has occurred, and further analysis is required.

For $ANC_{le,crit}$, many parameters were critical, including BC_w base rate, soil depth, growth rate, temperature, stem wood density, BC_w percent, and BC content in stems (Fig. 4C, Table 4). The most important parameters to sensitivity of $ANC_{le,crit}$ were BC_w base rate, soil depth, growth rate, stem wood density, and BC_w percent. A 20% increase in each of these parameters led to over a 90% increase in $ANC_{le,crit}$ (Table 4). This relationship may exist because BC_w base rate, soil depth, and BC_w percent control BC_w (Eqs. 3 and 5), while growth rate and stem wood density dominate BC_u (Eq. 4). The most significant parameters to uncertainty of CAL were BC_w base rate (57%), soil depth (18%), temperature (10%), and growth rate (9%), respectively (Table 4). $ANC_{le,crit}$ was also

Table 5
Effects of using extreme values of BC:Al ratio (i.e., the fourth simulation analysis) on estimation of CAL and $ANC_{le,crit}$

Target parameter	E_X (%)		
	CAL extreme BC:Al	ANC extreme BC:Al	CAL probabilistic BC:Al
BC_w base rate	47.93	44.91	62.22
BC:Al ratio	28.56	28.35	1.65
Soil depth	13.70	12.58	19.89
Temperature	6.81	6.20	10.89
BC content in stem	1.68	1.40	2.12
Growth rate	0.86	6.27	0.32
N content in stem	0.46		1.08
BC_w percent	0.29	0.29	0.10

The uncertainty measure, E_X, was the same as those described in Table 3. The significant change was that BC:Al ratio became the second critical factor in the uncertainty ranking for both CAL and $ANC_{le,crit}$, as compared to its ignorable rankings from simulations with probabilistic samples shown in Fig. 3 (included here in the third column) and Table 4.

examined to determine the effects of the two key parameters that define the chemical criterion of CAL: BC:Al ratio and Gibbsite constant. The results showed that BC:Al ratio exerted considerable influence on $ANC_{le,crit}$ and its aluminum component, [Al^{3+}], whereas Gibbsite constant had little effects on $ANC_{le,crit}$ and its hydrogen ion component, [H^+] (Fig. 5). Most effects of BC:Al ratio on $ANC_{le,crit}$ occurred at the lower end of its range.

4.4. Implications of observed behaviors of SMBE in calculating CAL

The results discussed above should have important implications to scaling up CAL with SMBE. Uncertainty in CAL estimates may be reduced by focusing on the most critical factors in the model. On the component level, BC_w and $ANC_{le,crit}$ should be the main focuses for potential reduction in CAL uncertainty. Future research should be directed toward efforts of quantifying BC_w given that the most critical parameters to $ANC_{le,crit}$ were also the three parameters of BC_w. The different methods of calculating BC_w described by Hodson and Langan (1999), Hall et al. (2001) and Gregor et al. (2004) should provide the starting points for such efforts. On the individual parameter level, significant improvement of CAL estimates should come from the three most influential parameters of BC_w base rate, soil depth, and temperature because they seem to exert the largest influence on the variability of CAL (Fig. 3). In addition, BC_w base rate must be represented by BC_w class as a function of soil properties in the scaling up process (Eq. 2) because it is difficult to define BC_w base rate directly in large scale assessment of CAL. Thus, the relationship between BC_w class and soil properties must be examined for uncertainty. Scaling up SMBE for national assessments of CAL should be greatly improved if variability and uncertainty in BC_w class, soil depth, and air temperature could be reduced in the spatial databases.

However, caution should be exercised because it is conceivable that the observed patterns of uncertainty may change under different circumstances (e.g., changes in the parameter space). Our preliminary analyses suggested that patterns of uncertainty values were relatively constant. We performed similar simulations analyses with extreme values of the parameters (i.e., use of minimum, average, and maximum values). This change of the parameter space was equivalent to increasing the CV of a parameter. For example, CV for BC:Al ratio and N_i respectively changed from 30% and 25% under the probabilistic sampling approach to 82% and 66% under the extreme value approach. We found that none of the parameters showed significant changes in their uncertainty rankings, except for BC:Al ratio. Using extreme values only for BC:Al ratio, we observed that BC:Al ratio became the second most important parameter to uncertainty of CAL and $ANC_{le,crit}$ (Table 5). Given that BC:Al ratio is the only environmental factor in SMBE that defines potential damages to ecosystems (Hodson and Langan, 1999) and that its "extreme" values are commonly used in Europe (1.0; Sverdrup and De Vries, 1994; Hall et al., 2001) and Canada (10.0; Watmough et al., 2004; Ouimet et al., 2006), BC:Al ratio should be treated as one of the most important parameters in SMBE calculations of CAL, especially when its values are low (Fig. 5). In addition, it is highly unlikely that a single value of BC:Al ratio could be sufficient for a large region in the scaling up of SMBE (Hall et al., 2001). Future research should establish relationships of BC:Al ratio to other ecosystem properties that can be easily defined at large scales and, thus, incorporated into scaling.

The results of this study demonstrate that the totality of a model must be emphasized and examined whenever possible because SMBE shows complex behaviors that could make results from partial analyses misleading. For example, BC:Al ratio changed significantly in its uncertainty ranking, but remained inconsequential in its sensitivity ranking, when extreme values were used (Table 5). BC:Al ratio and Gibbsite constant were both key biogeochemical parameters to $ANC_{le,crit}$. However, BC:Al ratio could be a major factor to the uncertainty of $ANC_{le,crit}$, whereas Gibbsite constant showed little effects on $ANC_{le,crit}$ (Tables 4 and 5, Fig. 5). In addition, $ANC_{le,crit}$ was highly sensitive to BC_w percent (P_{le}; Eq. 5), a factor that could be easily overlooked because of its lack of strong biogeochemical meaning (Table 4). If parameters like P_{le} can be defined in space (e.g., as a function of soil type), then they should be explicitly considered in uncertainty analysis and incorporated into the scaling of CAL. For reasons not quite clear, BC_u represents another example of the complex nature of uncertainty behaviors in SMBE. BC and N contents in stems were relatively important factors to CAL estimates (Fig. 3), but growth rate and stem wood density were the primary factors to BC_u and N_u (Table 4). Thus, it is imperative to examine models as a whole and not simply to infer model sensitivity from results obtained for the individual components, even though the models under examination are as simple as SMBE.

Future research should focus on effective ways of reducing uncertainty not only in SMBE, but also in the process of scaling up SMBE for national assessments of CAL. To ensure an acceptable level of uncertainty in model predictions is a major criterion that defines the adequacy of models and scaling algorithms (Li and Wu, 2006). In addition to the data uncertainty of natural variability in SMBE examined in this study, new sources of spatial uncertainty will come into play in the scaling up process, such as spatial heterogeneity of ecosystem properties at large scales and data quality of spatial databases. First, spatial uncertainty is manifested in categorical variables because of spatial heterogeneity within grid cells or pixels (Li and Wu, 2006). As a fundamental characteristic of ecological systems at all scales, spatial heterogeneity exerts significant influences on sampling, analysis, and modeling (Risser et al., 1984; Wiens, 1989; Li and Reynolds, 1995; Turner et al., 2001; Li and Wu, 2006). Spatial heterogeneity may be well represented by maps from spatial databases that define model input and parameters across the entire study area in a spatially explicit fashion. However, the within-cell or subpixel spatial heterogeneity is mostly ignored in most modeling exercises. For example, the certain existence of heterogeneity in soil texture in 1 km^2 cell commonly used for scaling will generate high uncertainty in BC_w base rate or BC_w class. Incorporating subpixel spatial heterogeneity of soil or ecosystem properties directly into scaling poses great difficulty. The associated spatial uncertainty should be quantified with data from intensively studied sites to assess its effects on the national assessments of CAL. Second, spatial uncertainty can be caused by poor data quality of model parameters defined by spatial databases in terms of errors in sampling, interpolation, and database management. Errors from these sources are bound to propagate through the scaling process. The spatial uncertainty from these error sources of spatial databases should be quantified to identify potential ways of reducing such error in the national assessments of CAL.

5. Conclusions

The results of this study strongly suggest that comprehensive uncertainty analysis of models should be performed to determine the most critical factors to sensitivity and uncertainty of their predictions. Our approaches in this study were systematic and comprehensive in that all factors in SMBE were considered and their effects on both CAL and its key components examined. The probability-based sampling was used to define the simulation parameter space. Advanced techniques of Monte Carlo simulation and uncertainty measures were applied to provide quantitative information about relative contributions of the parameters to the total uncertainty in CAL estimates. As a result, we were able to provide insights into the complex behaviors of SMBE prediction of CAL. The results of this study have:

1. indicated moderate uncertainty of CAL as predicted by SMBE with SD of 770 eq ha^{-1} yr^{-1} and CV of 40% (Table 2);
2. demonstrated quantitatively the dominance by BC_w and $ANC_{le,crit}$ in SMBE (Table 3);
3. identified the six most critical parameters to CAL: BC_w base rate (thus, BC_w class), BC:AL ratio, soil depth, temperature, BC content in stems, and N content in stems (Fig. 3, Table 5);
4. revealed the log-normal distribution of CAL with the probabilistic sampling (Figs. 1 and 2); and
5. most importantly, illustrated effective ways of error partition and uncertainty quantification of CAL.

These findings should prove useful to any application of SMBE to assess potential risks of air pollutants to ecosystems. Scaling up SMBE for national assessments of CAL with acceptable uncertainty is a major challenge for future research (Hodson and Langan, 1999; Barkman and Alveteg, 2001; Hall et al., 2001; Skeffington, 2006). Uncertainty analysis should play a critical role in ensuring that models provide sound and reliable scientific information required to develop effective policies for environmental protection.

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